

# **CERTIFICATE OF VERIFICATION**

I, Kwang Yeon HWANG of 648-23 Yeoksam-dong, Gangnam-gu, Seoul, Republic of Korea state that the attached document is a true and complete translation to the best of may knowledge of the Korean-English language and that the writings contained in the following pages are correct English translation of the specification and claims of the Korean Patent Application No. 10-2003-0020468.

Dated this 26th day of April, 2006.

Signature of translator:

Kwang Yeon HWANG



# KOREAN INTELLECTUAL PROPERTY OFFICE

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Application Number: Patent Application No. 10-2003-0020468

Date of Application: April 1, 2003

Applicant(s): LG Electronics Inc.

# **COMMISIONER**

### [ABSTRACT OF THE DISCLOSURE]

# [ABSTRACT]

Disclosed is an organic electroluminescent device including a substrate, a first and second electrode formed on the substrate, and a light-emitting layer formed between the first electrode and the second electrode. The light-emitting layer includes a plurality of materials which is a green luminescent material using a following chemical formula.

#### [Chemical formula 1]

In this case, at least one of A1 and A2 is selected from a substituted or non-substituted aromatic group, a heterocyclic group, an aliphatic group, and hydrogen.

# [INDEX WORDS]

green luminescent material, organic electroluminescent device

### [SPECIFICATION]

#### [TITLE OF THE INVENTION]

ORGANIC ELECTROLUMINESCENT DEVICE

#### [DETAILED DESCRIPTION OF THE INVENTION]

#### [OBJECT OF THE INVENTION]

# [FIELD OF THE INVENTION AND DISCUSSION OF THE RELATED ART]

The present invention relates to an organic electroluminescent device, and more particularly, a green luminescent material represented by a chemical formula below of a luminescent layer.

Recently, with the trend of a large sized display, a request of a flat display that occupies a small area has been increased. One example of the flat display is an organic electroluminescent device also called as an organic light emitting diode (OLED). And, technology for the organic electroluminescent display is developed rapidly, whereby various prototypes have been in market already.

The organic electroluminescent device emits light in a manner that electric charges are injected in an organic layer formed between an anode and a cathode so as to form a pair of electron and hole to generate an exciton and an excited state of the exciton falls to a ground state so as to emit light.

3

The organic electroluminescent device is not only formed on a flexible transparent substrate such as a plastic but also operated at a lower voltage (less than 10V) compared to a plasma display panel or an inorganic electroluminescent display.

Also, the organic electroluminescent device has advantages in that power consumption is reduced and various colors are available.

Moreover, the organic electroluminescent device enables to express three colors including green, blue, and red. Therefore, many concerns are focused on the organic electroluminescent device as the next generation full color display.

The organic electroluminescent device may be realized to emit blue, green, or red light in accordance with a material forming the luminescent layer.

Particularly, the light-emitting layer for green luminescence is fabricated by doping a dopant to a host material.

Perylene, coumarine, pyrene, anthracene, and a complexing agent such as Alq<sub>3</sub>, that is derivatives thereof are normally used.

However, it is a biggest problem in realizing green luminescence of the organic electroluminescent device that a degree of color purity and the luminescence efficiency are lowered.

The conventional green luminescent materials had problems of emitting light with a long wavelength during emission of light and of lowering the degree of color purity and the luminescence efficiency more at higher doping density.

Accordingly, the present invention is directed to an organic electroluminescent device that substantially obviates one or more problems due to limitations and disadvantages of the related art.

#### [TECHNICAL TASKS TO BE ACHIEVED BY THE INVENTION]

An object of the present invention is to provide an organic electroluminescent device having improved characteristics of luminescence in terms of impression of color, efficiency, as so on.

#### [PREFERRED EMBODIMENTS OF THE INVENTION]

To achieve these objects and other advantages and in accordance with the purpose of the invention, as embodied and broadly described herein, an organic luminescent device according to the present invention includes a substrate; a first and second electrodes formed on the substrate; a light-emitting layer formed between the first electrode and the second electrode, the light-emitting layer having a green luminescent material represented by a chemical formula 1.

## [Chemical formula 1]

In this case, at least one of A1 and A2 is selected from a substituted or non-substituted aromatic group, a heterocyclic group, an aliphatic group, and hydrogen.

The light-emitting layer comprises at least 2 materials including the green luminescent material of the chemical formula 1 and wt. % of the material in the chemical formula 1 is 0.1 - 90.9wt. % of a total weight of the luminescent layer.

Material forming the light-emitting layer together with the material of the chemical formula 1 is represented by a following chemical formula 2.

[Chemical formula 2]

B1 - X - B2

Wherein, X is fused Aromatic compounds and at least one of B1 and B2 is selected from a group consisting of aryl, alkylaryl, alkoxyaryl, arylaminoaryl, alkylamino, and arylallyl.

X is one selected from a group consisting of naphthalene, anthracene, phenanthrene, pyrene, perylene, and quinoline.

At least one of the B1 and B2 is selected from phenyl, biphenyl, pyridyl, naphthyl, tritylphenyl, biphenylenyl, anthryl, phenanthryl, pyrenyl, perylenyl, quinolyl, isoquinolyl, fluorenyl, terphenyl, tolyl, xylyl, methylnaphthyl, and hydrogen.

At least one of the A1 and A2 is selected from phenyl, biphenyl, pyridyl, naphthyl, quinolyl, isoquinolyl, fluorenyl, terphenyl, methyl, ethyl, propyl, i-propyl, and t-buthyl.

The substituent of each substituted A1 and A2 is at least one and selected from alkyl, aryl, alkoxy, alkylamino, halogen, aryloxy, arylamino, alkylsilyl, arylsilyl and hydrogen.

The substituent is one selected from methyl, ethyl, propyl, i-propyl, t-butyl, cyclohexyl, methoxy, ethoxy, propoxy, butoxy, dimethylamino, trimethylsilyl, fluorine, chroline, phenoxy, tolyloxy, dimethylamino, diethylamino, diphenylamino, and triphenylsilyl.

In the present invention, the light-emitting layer formed between a first electrode and a second electrode includes a plurality of materials and a green material using a following chemical formula as a dopant.

[Chemical formula 1]

In this case, at least one of A1 and A2 is selected from a substituted or non-substituted aromatic group, a heterocyclic group, an aliphatic group, and hydrogen.

Wt. % of the material in the chemical formula 1 is 0.1 - 49.9wt.% of a total weight of the luminescent layer.

Materials forming the light-emitting layer together with the material of the chemical formula 1 is structured as a chemical formula 2.

[Chemical formula 2]

In this case, the X is selected from fused aromatic compounds, particularly from a group consisting of naphthalene, fluorene, anthracene, phenanthrene, pyrene, perylene, quinoline, and isoquinoline.

Also, at least one of the B1 and B2 may be selected from a group consisting of aryl, alkylaryl, alkoxyaryl, arylaminoaryl, alkylamino, and arylallyl, particularly from phenyl, biphenyl, pyridyl, naphthyl, tritylphenyl, biphenylenyl, anthryl, phenanthryl, pyrenyl, perylenyl, quinolyl, isoquinolyl, fluorenyl, terphenyl, tolyl, xylyl, methylnaphthyl, and hydrogen.

In this case, the material forming the light-emitting layer together with the chemical formula 1 is one of following chemical formulas.

$$H-1$$
 $H-2$ 
 $H-3$ 
 $H-4$ 
 $H-5$ 
 $H-6$ 
 $H-7$ 
 $H-9$ 

$$H-28$$
 $H-29$ 
 $H-30$ 
 $H-30$ 

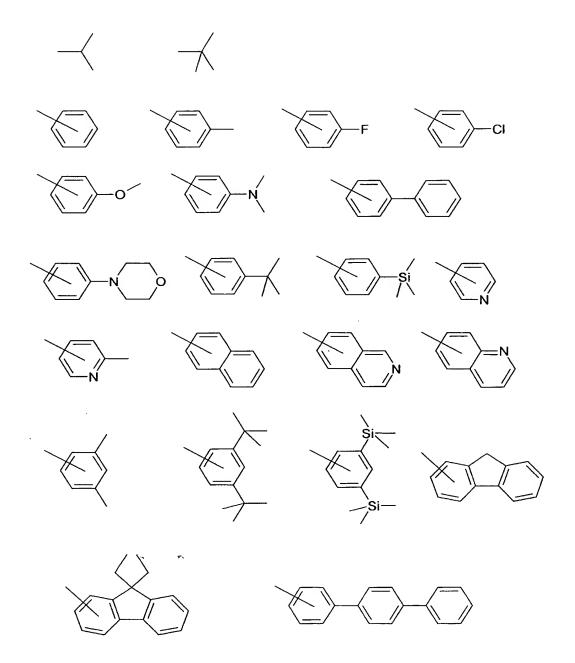
Also, in a case of the chemical formula 1, at least one of the A1 and A2 may be selected from a substituted or non-substituted aromatic group, a heterocyclic group, an aliphatic group, and hydrogen.

Particularly, at least one of A1 and A2 may be selected from phenyl, biphenyl, pyridyl, naphthyl, quinolyl, isoquinolyl, fluorenyl, terphenyl, methyl, ethyl, propyl, i-propyl, and t-buthyl.

In this case, each substitute of the substituted A1 and A2 is at lest one and selected from alkyl, aryl, alkoxy, alkylamino, halogen, aryloxy, arylamino, alkylsilyl, arylsilyl, and hydrogen.

For example, a substituent of the A1 and A2 may be selected from methyl, ethyl, propyl, i-propyl, t-butyl, cyclohexyl, methoxy, ethoxy, propoxy, butoxy, dimethylamino, trimethylsilyl, fluorine, chlorine, phenoxy, tolyloxy, dimethylamino, diethylamino, diphenylamino, and triphenylsilyl.

Particularly, at least one of the substituted or non-substituted A1 and A2 may be one of the following chemical formulas and the substituent of A1 and A2 is selected from a functional group having a following structural formula.



The green luminescent material may be at least one of the following chemical formulas.

$$G-1$$

$$G-2$$

$$G-3$$

$$G-4$$

$$G-6$$

$$G-6$$

$$G-7$$

$$G-8$$

$$G-15$$
 $G-16$ 
 $G-16$ 
 $G-16$ 
 $G-18$ 
 $G-19$ 
 $G-20$ 
 $G-21$ 
 $G-22$ 

$$G-23$$
 $G-24$ 

$$G-25$$

$$G-26$$

$$G-27$$

$$G-28$$

$$G-29$$
 $G-30$ 
 $G-30$ 
 $G-30$ 
 $G-30$ 
 $G-30$ 
 $G-30$ 

$$G-35$$
 $G-36$ 

$$G-36$$

$$G-36$$

$$G-36$$

$$G-36$$

$$G-36$$

$$G-36$$

$$G-36$$

$$G-38$$

$$G-38$$

$$G-38$$

$$G-39$$

$$G-40$$

$$G-41$$
 $G-42$ 
 $G-43$ 

According to the organic luminescent device of the present invention, N,N'-Di-naphthalen- 2-yl-N,N'-di-p-tolyl-anthracene-9,10-diamine used as the green luminescent material is compounded as follows.

# (1) Synthesis of N,N,N',N'-Tetraphenyl- anthracene-9,10-diamine

First, 3g(0.0089 mol) of 9,10-dibrmoanthracene, 3.78g (0.022 mol) of diphenylamine, 0.055g (1% mol) of BINAP [(2.2'-Bis(diphenylphosphino)-1,1'-

binaphthyl)], 0.02g (1% mol), Pd (OAc)<sub>2</sub> [Pd(II)acetate], (3.4g, 0.036mol) of NaO<sup>t</sup>Bu (Sodium tert-butoxide), and 100ml of toluene are admitted into a 2-necks-r.b.f. so as to be refluxed for 24 hours.

When the reaction is finished, the 2-necks-r.b.f. is cooled at a normal temperature and about 60ml of toluene which is a reaction solvent is removed by distillation under reduced pressure.

When methanol (100ml) is added to the solution from which toluene 40ml is removed, precipitate is obtained. And, the precipitate is filtered, whereby a yellow solid matter of N,N,N',N'-Tetraphenyl- anthracene-9,10-diamine is obtained.

# (2) Synthesis of N,N'-Diphenyl-N,N'- di-m-tolyl-anthracene-9,10-diamine

First, 3g(0.0089 mol) of 9,10-dibrmoanthracene, 5.21g (0.022 mol) of phenyl-m-tolyl-amine, 0.052g (1% mol) of BINAP, 0.02g (1% mol) of Pd (OAc)<sub>2</sub> and 3.4g (0.036mol) of NaO<sup>t</sup>Bu, and 100ml of toluene are admitted into a 2-necks-r.b.f. so as to be refluxed for 24 hours.

When the reaction is finished, the 2-necks-r.b.f. is cooled at a normal temperature and about 60ml of toluene which is a reaction solvent is removed by distillation under reduced pressure.

A resulted solution is extracted from a crude product from which toluene is removed using water and methanol (100ml). Methylene chloride layer is separated and removed from 'the resulted solution using MgSO<sub>4</sub> so as to remove methylene chloride by distillation under reduced pressure.

And, methanol is added thereto so as to clean the solid matter obtained by removing methylene chloride. And, the solid matter is filtered, whereby a yellow solid matter of N,N'-Diphenyl-N,N'- di-m-tolyl-anthracene-9,10-diamine is obtained.

# (3) Synthesis of N,N'-Di- naphthalen-2-yl-N,N'-di-p-tolyl-anthracene-9,10-diamine

First, 3g(0.0089 mol) of 9,10-dibrmoanthracene, 5.21g (0.022 mol) of Naphthalene-2-nyl-p-tolyl-amine, 0.052g (1% mol) of BINAP, 0.02g (1% mol) of Pd (OAc)<sub>2</sub> and 3.4g (0.036mol) of NaO<sup>t</sup>Bu, and 100ml of toluene are admitted into a 2-necks-r.b.f. so as to be refluxed for 24 hours.

When the reaction is finished, the 2-necks-r.b.f. is cooled at a normal temperature and about 60ml of toluene which is a 'reaction solvent is removed by distillation under reduced pressure.

A resulted solution is extracted from a crude product from which toluene is removed using water and methanol (100ml). Methylene chloride layer is separated

and removed from 'the resulted solution using MgSO<sub>4</sub> so as to remove methylene chloride by distillation under reduced pressure.

Methanol is added thereto so as to clean a solid matter obtained by removing methylene chloride. And, the solid matter is filtered, whereby a yellow solid matter of N,N'-Diphenyl-N,N'- di-*m*-tolyl-anthracene-9,10-diamine is obtained.

Hereinafter, preferred embodiments of the organic electroluminescent device will be described in accordance with the present invention.

### [Embodiment]

An ITO glass is patterned so as to have a size of 3mm x 3mm. The patterned ITO glass is then cleaned.

A substrate is loaded on a vacuum chamber of which basic pressure is set up as  $1x10^{-6}$  torr and CuPC(200Å), NPB(500Å), a light-emitting layer (300Å), Alq<sub>3</sub>(500Å), LiF(5Å), and Al(1,000Å) are deposited successively on the ITO in order.

In this case, a first host of the light-emitting layer includes a following material and a mixture ratio between the host and impurities is 1:0.01.

HOST-1

#### [First Embodiment]

When about 1mA of electric current is flown by using dopant having a chemical formula of G-2, voltage is about 7.48v and brightness shows about 1527cd/m<sup>2</sup>, whereby CIE (Commision Internationale de L'Eclairage) x=0.220, y=0.555.

#### [Second Embodiment]

When about 1mA of electric current is flown by using dopant having a chemical formula of G-3, voltage is about 7. 12v and brightness shows about  $1445 \text{cd/m}^2$ , whereby CIE x=0.254, y=0.619.

# [Third Embodiment]

When about 1mA of electric current is flown by using dopant having a chemical formula of G-32, voltage is about 7. 74v and brightness shows about  $1441 \text{cd/m}^2$ , whereby CIE x=0.297, y=0.615.

In this case, a structural formula of CuPC, NPB, Alq3 is as follows.

# [EFFECT OF THE INVENTION]

Accordingly, the present invention is a green luminescent material and an organic electroluminescent device having green color purity and a high luminescent efficiency is obtained by using such material with high color purity according to the present invention

#### [WHAT IS CLAIMED IS]

1. An organic organic electroluminescent device, comprising:

a substrate;

First and second electrodes formed on the substrate;

a light-emitting layer formed between the first electrode and the second electrode, the light-emitting layer having a green luminescent material represented by a chemical formula 1:

#### [Chemical formula 1]

wherein, at least one of A1 and A2 is selected from a substituted or non-substituted aromatic group, a heterocyclic group, an aliphatic group and hydrogen.

- 2. The organic electroluminescent device of claim 1, wherein the light-emitting layer comprises at least 2 materials including the green luminescent material of the chemical formula 1 and wt. % of the material in the chemical formula 1 is 0.1 90.0 wt.% of a total weight of the luminescent layer.
- 3. The organic electroluminescent device of claim 2, wherein a material forming the light-emitting layer together with the material of the chemical formula 1 is represented by a chemical formula 2:

[Chemical formula 2]

B1 - X - B2

611

wherein, the X is selected from a group consisting of fused aromatic compounds and at least one of the B1 and B2 is selected from a group consisting of aryl, alkylaryl, alkoxyaryl, arylaminoaryl, alkylaminoaryl, and arylallyl.

- 4. The organic electroluminescent device of claim 3, wherein the X is selected form a group consisting of naphthalene, anthracene, phenanthrene, pyrene, perylene, and quinoline.
- 5. The organic electroluminescent device of claim 3, wherein and at least one of the B1 and B2 is selected from phenyl, biphenyl, pyridyl, naphthyl, tritylphenyl, biphenylenyl, anthryl, phenanthryl, pyrenyl, perylenyl, quinolyl, isoquinolyl, fluorenyl, terphenyl, tolyl, xylyl, methylnaphthyl, and hydrogen.
- 6. The organic electroluminescent device of claim 3, wherein the material forming the light-emitting layer together with the material of the chemical formula 1 is one of following formulas:

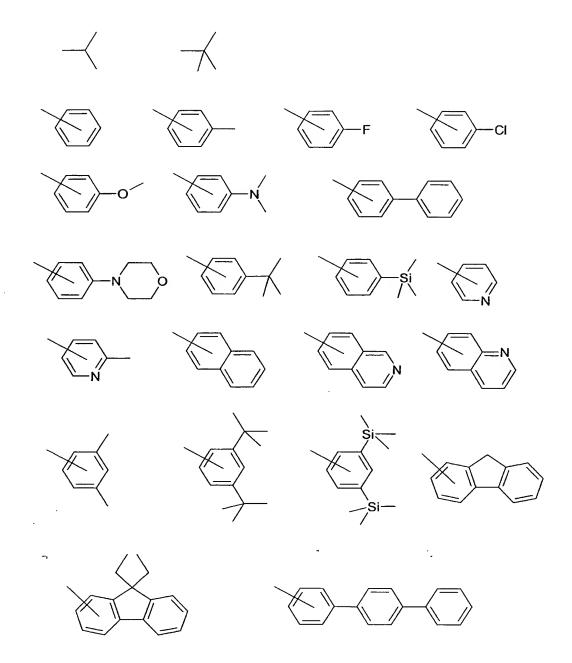
$$H-1$$
  $H-2$   $H-3$   $H-9$   $H-9$   $H-9$ 

$$H-28$$
 $H-29$ 
 $H-30$ 
 $H-30$ 
 $H-30$ 

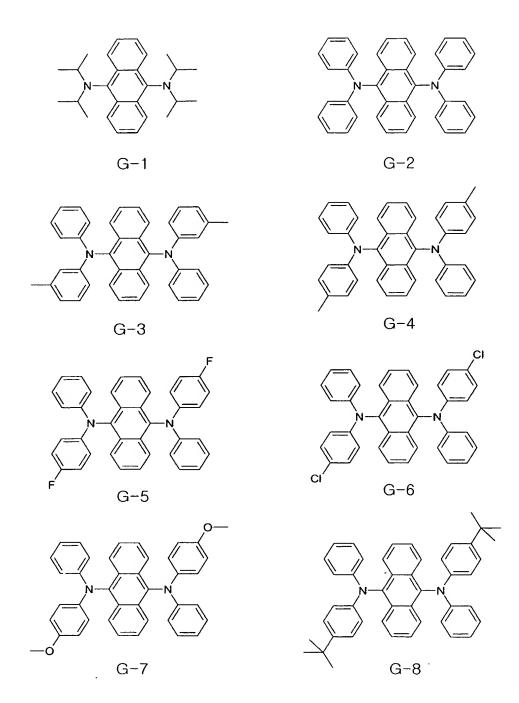
- 7. The organic electroluminescent device of claim 1, wherein at least one of the A1 and A2 is selected from phenyl, biphenyl, pyridyl, naphthyl, quinolyl, isoquinolyl, fluorenyl, terphenyl, methyl, ethyl, propyl, i-propyl, and t-buthyl.
  - 8. The organic electroluminescent device of claim 1, wherein a substituent of each substituted A1 and A2 is at least one and selected from alkyl, aryl, alkoxy, alkylamino, halogen, aryloxy, arylamino, alkylsilyl, arylsilyl and hydrogen.
    - 9. The organic electroluminescent device of claim 8, wherein the substituent is

one selected from methyl, ethyl, propyl, i-propyl, t-butyl, cyclohexyl, methoxy, ethoxy, propoxy, butoxy, dimethylamino, trimethylsilyl, fluorine, chroline, phenoxy, tolyloxy, dimethylamino, diethylamino, diphenylamino, and triphenylsilyl.

10. The organic electroluminescent device of claim 1, wherein at least one of the A1 and A2 in one of following chemical formulas.



11. The organic electroluminescent device of claim 1, wherein the green luminescent material is at least one of following chemical formulas:



$$G-15$$
 $G-16$ 
 $G-16$ 
 $G-16$ 
 $G-18$ 
 $G-18$ 
 $G-19$ 
 $G-20$ 
 $G-21$ 
 $G-22$ 

$$G-23$$
 $G-24$ 

$$G-25$$

$$G-26$$

$$G-27$$

$$G-28$$

$$G$$
-29  $G$ -30  $G$ -31  $G$ -32  $G$ -32

G-33

G-34

G-35

3,

